

## Biophysical and economic limits to negative CO<sub>2</sub> emissions

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Figures: 4

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**Methods:**

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**To limit warming below 2°C with a >50% chance, most recent scenarios from integrated assessment models (IAMs) require large-scale deployment of negative emissions technologies (NETs), i.e. technologies that result in the net removal of greenhouse gases (GHGs) from the atmosphere. We quantify potential global impacts of the different NETs on land, GHG, water, albedo, nutrient, energy and costs, to determine the biophysical limits to, and economic costs of, the widespread application of NETs. Resource implications vary between technologies and need to be satisfactorily addressed if NETs are to a significant role in achieving climate goals.**

Despite two decades of effort to curb emissions of CO<sub>2</sub> and other greenhouse gases (GHGs), emissions grew faster during the 2000s than in the 1990s<sup>1</sup> and by 2010 had reached ~50 Gt CO<sub>2</sub>eq./yr<sup>2,3</sup>. The continuing rise in emissions is a growing challenge for meeting the international goal of limiting warming to less than 2°C relative to the preindustrial era, particularly without stringent climate policies to decrease emissions in the near future<sup>2-4</sup>. Since NETs now appear ever more necessary<sup>3,5-10</sup>, society needs to be informed of the potential risks and opportunities afforded by all mitigation options available, to decide which pathways are most desirable for dealing with climate change.

There are several distinct classes of NETs including: (1) Bioenergy (BE) with carbon capture and storage (CCS; together referred to as BECCS<sup>12,13</sup>), (2) direct air capture of CO<sub>2</sub> from ambient air by engineered chemical reactions (or DAC<sup>14,15</sup>), (3) enhanced weathering of minerals (or EW<sup>16</sup>) where natural weathering to remove CO<sub>2</sub> from the atmosphere is accelerated, and the products stored in soils, or buried in land/deep ocean<sup>17-20</sup>, (4) afforestation and reforestation (AR) to fix atmospheric carbon in biomass and soils<sup>21-23</sup>, (5) manipulation of uptake of carbon by the ocean either biologically (i.e. by fertilizing nutrient limited areas<sup>24,25</sup>) or chemically (i.e. by enhancing alkalinity<sup>26</sup>), (6) altered agricultural practices, such as increased carbon storage in soils<sup>27-29</sup>, and (7) converting biomass to recalcitrant biochar, for use as a soil amendment<sup>30</sup>. In this study, we focus on BECCS, DAC, EW and AR, since large uncertainties pertain to the ocean-based strategies (e.g. ocean iron fertilization<sup>31</sup>), and other land-based approaches (e.g. soil carbon / biochar storage) have been evaluated elsewhere<sup>32-34</sup>. Figure 1 depicts the main flows of carbon among atmospheric, land, ocean and geological reservoirs

for fossil fuel combustion (Fig. 1A), BE (Fig. 1B), CCS (Fig. 1C), and the altered carbon flows entailed by each NET (Fig. 1D-1H) when carbon is removed from the atmosphere.

[Figure 1 here]

Coupled energy-land-use analyses of NETs using IAMs have so-far focussed primarily on BECCS<sup>7,35,36</sup> and AR strategies<sup>37-40</sup>. These studies suggest that there may be considerable cost-competitive potential for these strategies. Other NET options have been studied<sup>14,20,41</sup>, but most IAMs do not yet represent them. Most IAMs allow biomass-based NETs in the production of electricity and heat in power stations as well as hydrogen generation, and sometimes for generating other transport fuels or bioplastics. The key distinguishing feature of NETs is their ability to remove CO<sub>2</sub> from the atmosphere. Depending on the development of overall emissions, this may lead to: a) a global net removal of CO<sub>2</sub> from the atmosphere by offsetting emissions that were released either in the past or in the near future<sup>42</sup>; or b) offsetting ongoing emissions from difficult-to-mitigate sources of CO<sub>2</sub> such as the transportation sector<sup>43,44</sup>, and non-CO<sub>2</sub> GHGs.

The IPCC AR5 scenario database includes 116 scenarios consistent with >66% probability of limiting warming below 2°C (i.e. with concentration levels of 430–480 ppm CO<sub>2</sub>-eq. in 2100)<sup>42</sup>. Of these, 101 (87%) apply global NETs in the second half of this century, as do many scenarios that allow CO<sub>2</sub> concentrations to grow between 480 and 720 ppm CO<sub>2</sub>-eq. by 2100 (501/653 apply BECCS; with 235/653 [36%] delivering net negative emissions globally<sup>42</sup>; see also Figure 2).

[Figure 2 here]

Results from two recent modelling exercises<sup>10,36,45</sup> show that median BECCS deployment of around 3.3 GtC/yr (Table S3) is observed for scenarios consistent with the <2°C target (430-480 ppm CO<sub>2</sub>eq.); we assess other NETs for deployment levels that give the same negative emissions in 2100 (Methods).

A key question is whether these rates of deployment of NETs can be achieved and sustained. Most of the NETs require use of land and water, some use fertiliser, and may also impact albedo. All

NETs are expected to have considerable costs<sup>8,10</sup>. Earlier studies have examined a number of constraints to NETs<sup>7,38-40,48-53</sup>, but have not assessed different the range of different NET types together, or considered the range of impacts included here. We perform a 'bottom up' implied resource use analysis rather than a 'top-down' potential efficacy analysis, using the best available data from the most recent literature to provide values for the analysis. The evidence base for the values used varies greatly between NETs, with some (e.g. BECCS) having been the subject of a large body of research, whilst others (e.g. EW) have received less attention. The data sources and a qualitative assessment of the confidence / uncertainty in the ranges we derive are described in detail in the Methods. We estimate the impacts of each NET per unit of negative emission, i.e. per-t-Ceq., then assess the global resource implications, focussing on the limits to large-scale NET deployment, and how these differ between NETs.

### **Impacts of NETs per unit of negative emissions**

NETs vary dramatically in terms of their requirements for land, GHG emissions removed or emitted, water, and nutrients, energy produced or demanded, biophysical climate impacts (represented by surface albedo) and cost, depending on both their character and on the scale of their deployment. Figure 3 highlights the differences in these requirements expressed per-t-Ceq. removed from the atmosphere. Geological storage capacity has recently been evaluated as a potential limit to implementation for CCS (and hence BECCS)<sup>54,55</sup>, so is not considered further here. Indirect effects of NETs through the reduced use of other technologies in pursuit of a given goal, e.g. potentially fewer nuclear reactors, wind farms, solar arrays etc., are not considered here. The values used are estimated from analyses presented in the latest peer-reviewed literature (Methods).

[Figure 3 here]

*Land area and GHG emissions:* The area (and type) of land required per unit of Ceq. removed from the atmosphere, also termed the land use intensity, is particularly important for land-based NETs (Fig. 3A). The land use intensity of BECCS is quite high, with values ranging from ~1-1.7 ha/tCeq./yr where forest residues are used as the BE feedstock, ~0.6 ha/ tCeq./yr for agricultural residues, and 0.1-0.4 ha/ tCeq./yr when purpose-grown energy crops are used (Table S2 column “Carbon in biomass available for capture” shows values in tCeq./ha/yr). Table S2 shows the carbon and GHG emissions / removals associated with a range of energy crops and forest types, and the net negative emissions delivered (Methods). EW and DAC have minimal land requirements with land use intensities of <0.01 ha/tCeq./yr<sup>19</sup> and <0.001 ha/tCeq./yr<sup>15</sup>, respectively (Fig. 3A).

*Water:* Water use between different BE feedstocks (including forest feedstocks) is highly variable and is generally considered to be higher for short rotation coppice and C4 grasses than for annual crops and grassland on an area basis<sup>56</sup>, though when corrected for biomass productivity, the ranges are closer and overlap considerably<sup>57</sup> (Fig. 3B). In calculating water implications of BECCS, water use for CCS is added to the BE water use (Methods). Where deployed, irrigation also has a dominant impact on water use. Estimates of water required per-t-Ceq. removed by DAC and EW are an order of magnitude or more lower than for BECCS (Fig. 3B). For EW of olivine, one molecule of water is required for each molecule of CO<sub>2</sub> removed, so each tCeq. would require 1.5 m<sup>3</sup> water (Fig. 3B).

*Energy:* Energy input / output varies considerably between NETs. BECCS has a positive net energy balance, with energy production ranges of 3-40 GJ/tCeq. for energy crops<sup>58</sup> (Fig. 3E). DAC and EW, on the other hand, require considerable energy input to deliver C removals, with the minimum theoretical energy input requirement for the chemical reactions only of DAC<sup>15</sup> of 1.8 GJ/tCeq. removed at atmospheric concentrations of CO<sub>2</sub>, and for EW of olivine of 0.28-0.75 GJ/tCeq. (Fig. 3E). When also including other energy inputs for mining, processing, transport, injection etc., the energy inputs for DAC and EW are much greater, perhaps as much as 45 GJ/tCeq. and 46 GJ/tCeq., respectively<sup>15,59</sup> (Fig. 3E). The GHG implication of this additional energy use depends on the GHG intensity of energy supply, which is likely to change over the rest of this century. Energy requirement

is less important if low carbon energy is used, but may still have additional impacts (e.g. *via* large area of solar PV panels to power DAC plants<sup>48</sup>).

*Nutrients:* Nutrients are depleted when biomass is removed from a field or ecosystem for use as a BE feedstock, so this is an issue for BECCS, for AR when biomass is removed from the site, but not for DAC or EW. Perennial energy crops typically contain around 10 kgN/tCeq. (and 0.8 kgP/tCeq. in the case of *Miscanthus*<sup>60</sup>), trees around 4-5 kgN/tCeq., and annual energy crops (such as fibre sorghum) around 20 kgN/tCeq. Nutrient removal therefore differs several fold among biomass sources (Fig. 3C), so large-scale transition to using land for biomass production could deplete nutrients, but this will depend on the vegetation / land use that is replaced. Additional nutrient requirements (i.e. fertilization) are difficult to estimate on a net basis, since fertilizer may also be used (more or less intensively) on the land use replaced by energy crops<sup>61</sup>. Nutrient depletion further translates into agricultural inputs and upstream GHG emissions and energy consumption.

*Albedo:* In addition to biogeochemical climate impacts (e.g. uptake of atmospheric carbon), changes in land use affect climate by altering the physical characteristics of the Earth's surface, such as increased evapotranspiration<sup>62</sup>, increased cloud cover in the tropics<sup>63</sup>. Important among these physical changes is albedo (surface albedo used here), which is the reflectance by the earth's surface. The albedo of lighter-coloured and less dense vegetation (e.g. food crops, grasses) is much greater than that of trees<sup>56,64</sup>. The situation is further complicated in areas where shorter vegetation may be persistently covered by highly-reflective snow in winter, while tall coniferous trees remain exposed and therefore much-less reflective<sup>64</sup>. This snow-mediated effect is large enough to mean that AR in northerly latitudes may have a neutral or net warming effect (larger than the carbon sink provided by the vegetation)<sup>65-68</sup>. Figure 3D shows the change in albedo under different NETs, focussing on the replacement of cropland or grassland with energy crops, or under AR, both with and without the snow effect on albedo.

*Costs:* The economic costs of deploying and operating NETs will vary according to the specific technologies involved, the scale of deployment and observed learning, the amount and value of co-

products, site-specific factors and the scale and cost of building and maintaining any supporting infrastructure (costs of capturing and storing a tCeq. are from studies using ~\$(US)\$<sub>2005</sub> to \$<sub>2015</sub> values). In the case of BECCS and DAC, costs can be anticipated to occur across three stages: (1) capture, (2) transport and (3) storage (including monitoring and verification). Recent estimates of the total costs of DAC technologies<sup>41,69</sup> are \$1,600-\$2,080 /tCeq., of which roughly two-thirds are capital costs and one-third operating costs (Fig. 3F). Though there are very wide ranges for costs of BECCS<sup>70</sup>, the mean price of BECCS estimated across 6 IAMs in 2100<sup>49</sup> was \$132/tCeq. (Fig. 3F); costs of bioenergy without CCS are lower<sup>57,58</sup>. AR costs are estimated to be \$65-108/tCeq. in 2100, with a mean of \$87/tCeq. Estimated costs of EW are taken from Renforth<sup>59</sup>: \$88-\$2,120/tCeq., around a mean of \$1,104/tCeq; these estimates are uncertain and the relative balance between capital and operating costs has not yet been thoroughly examined.

### **Global resource implications of NETs deployment**

We use global deployment of BECCS in the recent assessments featured in Table S3 to derive the corresponding resource implications (Table 1), and focus on the scenario giving a 2100 atmospheric CO<sub>2</sub> concentration in the range of 430-480 ppm (consistent with a 2°C target). We compare DAC resource implications at the same level of negative emission as BECCS (i.e. 3.3 GtCeq./yr in 2100; Table 1). For other NETs, which are not able to meet the same level of emissions removal, we use values compiled from an analysis of the recent literature to give mean and maximum implementation levels (Methods). Mean values for carbon removals from AR are estimated to be around 1.1 GtCeq./yr by 2100, with a maximum value of 3.3 GtCeq./yr with very large-scale deployment<sup>6,7,71</sup> (Table 1). The potential of carbon removal by EW (including both adding carbonate and olivine to oceans and soils) has been estimated to be as great as 1 GtCeq./yr by 2100, but with mean annual removal an order of magnitude less<sup>71</sup>: 0.2 GtCeq./yr. Combined with the bottom-up, per-t-Ceq. impact ranges (Methods), we then assess the resource implications, and the extent to which available resources may limit the deployment of NETs globally.



[Table 1 here]

*Land area:* DAC has a small direct land footprint (Fig. 3A) and can be deployed on unproductive land that supplies few ecosystem services<sup>15</sup>, though the land footprint could be considerable if solar PV panels or wind turbines were used to provide the energy required for DAC<sup>48</sup>. EW has a somewhat larger land footprint if the minerals are applied to the land surface (as opposed to the oceans, or if weathering reactions occur in industrial autoclaves), though crushed olivine or carbonates could be spread on agricultural and forest land to allow the weathering to take place, with co-benefits of raising the pH of acidic soils to make them more productive<sup>16</sup>. Thus, EW technologies may not always compete for land with other uses, despite the large areas involved (e.g. the estimated potential of 1 GtCeq./yr removed might require 10 Mha)<sup>16</sup>.

Assuming per-area carbon in biomass available for capture as a feedstock for BECCS of widely-applicable, high-productivity dedicated energy crops (Willow/Poplar SRC and Miscanthus; 4.7-8.6 tCeq./ha/yr; Table S2), BECCS delivering 3.3 GtCeq./yr of negative emissions would require a land area of ~380-700 Mha in 2100 (Table 2), with a wider possible range determined by productivity (Table S2). This emissions removal is equivalent to 21% of total current human appropriated NPP (15.6 GtC/yr in 2000<sup>72</sup>), or 4% of total global potential NPP<sup>72</sup>. Areas for AR calculated assuming a mean carbon uptake of AR over the growth period of 3.4 tCeq./ha/yr (Methods; Fig. 3A) gives a land area for AR corresponding to 1.1 and 3.3 GtC/yr removed in 2100 of ~320 and ~970 Mha, respectively, similar to other estimates<sup>53</sup>. Estimates of land use by BECCS and AR are consistent with the values presented in previous studies<sup>50</sup> for three IAMs (GCAM, IMAGE and ReMIND / MAgPIE), though other studies suggest larger areas<sup>40</sup>. Without global forest protection, increased BE deployment would increase GHG emissions from land-use change<sup>73</sup>.

Total agricultural land area in 2000 was ~4,960 Mha, with an area of arable and permanent crops of ~1,520 Mha<sup>74</sup>, so area for BECCS (380-700 Mha) represents 7-25% of agricultural land, and 25-46% of arable plus permanent crop area. AR (at 1.1-3.3 GtCeq./yr negative emissions; 320-970 Mha, respectively) represents 6-20% of total agricultural land, and 21-64% of arable plus permanent

crop area. This range of land demands are 2-4 times larger than land identified as abandoned or marginal<sup>75</sup>. Thus, use on large areas of productive land is expected to impact land available for food or other bioenergy production<sup>13,38,76-78</sup>, and the delivery of other ecosystem services<sup>13,33,79</sup>, which may prove to be a limit to implementation of BECCS<sup>80</sup> and AR. One uncertainty is the future rate of increase of food crop yields<sup>38,81</sup> and whether this will meet future food demand<sup>82</sup>, thereby potentially freeing more cropland for BECCS or AR, even if at higher price<sup>38</sup>.

*Water:* Water stress is increasing worldwide, attributable to rising water demands and reduced supplies, both of which can be exacerbated in some locations by climate change<sup>83</sup>. In particular, the evaporative demand of plants increases with temperature as vapour pressure deficit increases. Evaporative loss can be 20-30 mol H<sub>2</sub>O per mol CO<sub>2</sub> absorbed by an amine DAC unit<sup>15,84</sup>, giving a water use estimate of ~92 (73-110) m<sup>3</sup>/tCeq. (Fig. 3B). Implementation at levels of 3.3 GtCeq./yr in 2100 (Table 1) would therefore be expected to use ~300 km<sup>3</sup>/yr water assuming current amine technology, which is 4% of the total current evapotranspiration used for crop cultivation<sup>85</sup>. Sodium hydroxide for DAC, however, uses 3.7 m<sup>3</sup>/t Ceq.<sup>-1</sup> (Fig. 3B)<sup>84</sup>, so equivalent levels of implementation using sodium hydroxide in place of amines would result in water use of ~10 km<sup>3</sup>/yr. For EW, with a water use of 1.5 m<sup>3</sup>/tCeq. (Figure 3B), deployment to remove 0.2 (mean) or 1 (maximum) GtCeq./yr, would involve water use of 0.3 and 1.5 km<sup>3</sup>, respectively.

Water use for forests is estimated to be 1,765 (1,176-2,353) m<sup>3</sup>/tCeq./yr, which includes both interception and transpiration (Fig. 3B). However, since trees replace other vegetation during AR, the total net impact must be calculated by subtracting the water use of the previous land cover. Assuming a water use similar to short vegetation of 1,450 (900-2,000) m<sup>3</sup>/tCeq./yr prior to AR (Fig. 3B), the additional water use from AR is estimated to be around 315 m<sup>3</sup>/tCeq./yr, which is 1% of total evapotranspiration from current forests<sup>85</sup>. For AR delivering capture of 1.1 or 3.3 Gt C/yr (Table 1), additional water use is thus estimated to be ~370 km<sup>3</sup>/yr or 1,040 km<sup>3</sup>/yr, respectively.

Similar calculations can be made for BECCS. For unirrigated BE, evaporative loss is estimated to be 1,530 (1,176-1,822) m<sup>3</sup>/tCeq./yr (Fig. 3B), 80 m<sup>3</sup>/tCeq./yr more than for average short

vegetation (Fig. 3B). Thus, deployment of BECCS at 3.3 GtCeq./yr in 2100 would lead to additional water use from the crop production phase of  $\sim 260 \text{ km}^3/\text{yr}$ . There is an opportunity cost of using soil moisture for sequestration / BE production rather than for growing food. Our estimates for water use are an order of magnitude lower than other recent estimates for BE crops<sup>51</sup> and for AR<sup>53</sup>, since water use in those studies<sup>51,53</sup> were expressed as a *total* rather than *additional* water use due to land use change, and for BE<sup>51</sup> also considered irrigation. Since irrigated BE crops were estimated to double agricultural water withdrawals in the absence of explicit water protection policies<sup>51</sup>, which could pose a severe threat to freshwater ecosystems, as human water withdrawals are dominated by agriculture and already lead to ecosystem degradation and biodiversity loss. Land requirements for BE crops would greatly increase (by  $\sim 40\%$ , mainly from pastures and tropical forests) if irrigated BE production was excluded, meaning that there will be a trade-off between water and land requirements if BE is implemented at large scales<sup>51</sup>.

For BECCS, additional water is required for CCS, adding about  $450 \text{ m}^3/\text{tCeq.}/\text{yr}$  to the evaporative loss relative to BE alone (Fig. 3B<sup>15</sup>), equivalent to an additional water use due to BECCS of  $\sim 720 \text{ km}^3/\text{yr}$  (sum of additional evaporative loss plus CCS water use), for the 3.3 GtCeq./yr by 2100 level of implementation (Table 1). BECCS would thus require an additional quantity of water equivalent to  $\sim 10\%$  of the current evapotranspiration from all cropland areas worldwide<sup>85</sup>.

To put these figures in context, total global renewable freshwater supply on land<sup>86</sup> is  $110,300 \text{ km}^3/\text{yr}$  of which humans appropriate  $24,980 \text{ km}^3/\text{yr}$ <sup>86</sup>, so implementation of BECCS at 3.3 GtCeq./yr by 2100 represents an additional use of  $\sim 3\%$  of currently human appropriated freshwater. AR implemented at 1.1 GtCeq./yr by 2100 would represent 1-2% of human appropriated freshwater. Expressing additional water use as a proportion of runoff in a region would give a more accurate picture of the threat to water resources at a given location, but without a spatially disaggregated analysis this is not feasible. Nevertheless, with human pressures on freshwater increasing<sup>83,87</sup>, water use could act as a significant limitation to implementation of high-water-demand NETs, such as BECCS.

*Energy:* BE currently supplies about 10% of primary energy worldwide<sup>58</sup>, i.e. an estimated 44.5 EJ/yr. Of this, 74% comes from fuel wood, 9% from forest and agricultural residues, 8% from recovered wood, 6% from industrial organic residues, and 3% from dedicated energy crops<sup>58</sup>. Most of this biomass, however, could not currently be used for BECCS, as the vast majority is used in small scale applications, e.g. for household cooking and heating in developing countries<sup>58</sup>. BECCS delivering 3.3 GtCeq./yr of negative emissions would deliver ~170 EJ/yr of primary energy in 2100<sup>10,36,45</sup> (Table 1). Estimates of future energy potential vary greatly; there is high consensus that 100 EJ/yr could be attained, medium agreement that 100-300 EJ/yr could be attained, but there is low consensus that primary energy above 300 EJ/yr could be supplied by BE<sup>13,33</sup>. Stabilization scenarios from the IAM literature suggest that BE could supply from 10 to 245 EJ/yr of global primary energy by 2050<sup>73,88</sup>, and deliver a sizable contribution to primary energy in 2100<sup>42</sup>.

The energy required by AR is very low (site preparation only) and is assumed here to be negligible. Other NETs have large energy demands (Fig. 3E). Using our realistic estimate of 46 GJ of energy required per-t -Ceq. removed by EW (Fig. 3), the 0.2 to 1.0 Gt C/yr that might be captured (Table S2) would entail up to 46 EJ/yr of energy in 2100 (Table 1). The energy requirements of amine DAC<sup>15</sup> (Fig. 3E) deployed for net removal of ~3.3 GtCeq./yr would amount to a global energy requirement of 156 EJ/yr if all energy costs are included (Table 1). This is equivalent to 29% of total global energy use in 2013 (540 EJ/yr), and a significant proportion of total energy demand in 2100 (IPCC AR5 scenario database: ~500-1,500 EJ/yr), which will be a major limitation unless low-GHG energy could be used, or the energy requirements significantly reduced.

*Nutrients:* DAC has no impact on soil nutrients, and EW may, in some cases, provide beneficial minerals and pH adjustment that are difficult to quantify at aggregate level. Nutrient concentrations in crop biomass are often higher than in tree biomass (Fig. 3C), but nutrients are removed from cropland and grazing land in agricultural products, while AR on agricultural land is likely to increase the retention of nutrients within an ecosystem. However, nutrient limitation could limit productivity, which may limit carbon storage<sup>52</sup>. Nutrients are also removed when BE feedstocks are removed from

the site on which they are grown, resulting in depletion of nutrients relative to land uses where biomass is not removed, but not necessarily at the same level as agricultural land<sup>89</sup>. BE feedstocks with low nutrient concentrations, such as residue-, forest- and lignocellulosic-biomass should hence be favoured over feedstocks with higher nutrient concentrations. Assuming nutrient concentrations of forests of 2.0 to 5.1 kg N/tCeq. (Fig. 3C), and that most nutrients are removed at harvest for energy and food crops, AR areas of ~320 and 970 Mha, consistent with AR removing 1.1 (mean) and 3.3 (high) GtCeq./yr (Table 1), would increase global nitrogen retention in biomass by 2.2-5.6 and 6.6-16.8 kt N/yr, respectively. Scaling values for implementation of 1 GtCeq./yr of negative emissions<sup>53</sup>, P and N demand to balance to carbon stored is estimated to be 220-990 kt P/yr and 100-1,000 kt N/yr for AR at 1.1-3.3 GtCeq./yr of negative emissions, though these values are absolute, and do not account for the P and N in the vegetation replaced by AR.

*Albedo:* The effect of DAC and EW on the reflectivity of the earth surface is assumed to be small (excluding possible use of solar PV to generate energy for DAC<sup>48</sup>; Fig. 3D). However, the land areas required for BECCS and AR can dramatically affect albedo (Fig. 3D). Since the effect is greatly amplified by the presence of snow, the exact location of the BECCS or AR (latitude and elevation), and the vegetation it replaces, is critical in assessing the impact on albedo (Fig. 3D). Albedo can significantly reduce<sup>65</sup> or even reverse net radiative forcing from AR at northern latitudes<sup>66</sup>. This observation could limit the value of AR for climate mitigation in northerly regions. For BECCS, replacement of short vegetation with taller vegetation (e.g. Miscanthus, SRC), could have similar effects on albedo, though likely less than the impact of AR with coniferous forest (Fig. 3D). Because AR is more likely to occur at high latitudes than production of BECCS feedstocks, BECCS should not have a deleterious impact on albedo. At low to mid-latitudes, AR could increase radiative forcing by decreasing albedo, but without a regional distribution, the scale of these impacts cannot be assessed.

*Investment needs:* The deployment of NETs (specifically BECCS) in IAM scenarios is an outcome of an optimization of costs over time. The existence of large-scale gross negative emissions even in less ambitious stabilization pathways indicates that BECCS is selected as a cost-effective component of the energy mix, allowing higher residual emissions elsewhere, which would otherwise be more

expensive to abate. Investments into BECCS provide an additional indicator for assessing the scale and speed of BECCS deployment over the next several decades. Table S4 summarizes investment estimates from six global integrated assessment models that assessed 2°C scenarios within the context of the LIMITS model inter-comparison<sup>90</sup> for 2030 and 2050: \$36.2 and \$29.4 billion/yr worth of investment is estimated as optimal by 2030 for scaling up biomass electricity and biofuels production technologies worldwide on average, respectively. By 2050, these investment levels grow to \$138.3 and \$122.6 billion/yr, respectively<sup>90</sup> (Table S4). This represents 5 and 4%, respectively, of projected total global energy system investments required by 2050 of \$2,932 (inter-model range: \$1,889 – \$4,338) billion/yr<sup>90</sup>. Investment needs for DAC, EW and AR are not known, but given the much higher unit costs (per-t-Ceq.) for DAC, the higher costs of EW and the lower unit costs of AR described in section 2, the investment needs are estimated qualitatively (relative to BECCS; Table 1).

[Figure 4 here]

The aggregate impacts of NETs on land, energy, water, and relative investment needs for levels of implementation equivalent to BECCS in scenarios consistent with a 2°C target (3.3 Gt Ceq./yr or mean and maximum attainable where that level of negative emissions cannot be reached) described in this section, are summarised schematically in Figure 4.

## Discussion

Biophysical, biogeochemical (nutrients), energy and economic resource implications of large-scale implementation of NETs differ widely among NETs. For DAC, costs and energy requirements are currently prohibitive and can be anticipated to slow deployment. R&D is needed to reduce costs and energy requirements. For EW, the land areas required for spreading and/or burying crushed olivine are large, such that the logistical costs may represent an important barrier, compounded by the fact that the plausible potential for carbon removal is lower than for other NETs. In contrast, AR is relatively inexpensive, but the unintended impacts on radiative forcing *via* decreased albedo at high

latitudes and increased evapotranspiration increasing the atmospheric water vapour content, could limit effectiveness; likewise increased water requirements could be an important trade-off, particularly in dry regions. Competition for land is also a potential issue, as it is for BECCS<sup>53,91,92</sup>. BECCS may also be limited by increased water use, particularly if feedstocks are irrigated and when the additional water required for CCS is considered, or by nutrient demand. These biophysical and economic resource implications may directly impose limits on the implementation of NETs in the future, but they may also indirectly constrain NETs by interacting with a number of societal challenges facing humanity in the coming decades, such as food, water and energy security, and thereby sustainable development. In addition to the biophysical and economic limits to NETs considered here, social, educational and institutional barriers, such as public acceptance of, and safety concerns about, new technologies and related deployment policies, could limit implementation. The drivers, risks, and limitations of the supply of NETs, showing activities thought to increase the potential supply of NETs, as well as the risks and geophysical/societal limits to the potential of NETs are shown in Figure S1. Commercialisation and deployment at larger scales also allows learning, efficiency improvement and cost reduction.

To inform society of the potential risks and opportunities afforded by all mitigation options available, more research on NETs is clearly required. Though we have collated the best available data on NET impacts and have reflected changes related to deployment scale as accurately as possible, it is clear that common modelling frameworks are required to implement learning, cost, supply and efficiency curves for all NETs. By implementing such curves for all NETs, future models will be able to develop portfolios of trajectories of NET development, allowing least cost options to be selected, and learning / efficiency improvements to be reflected. The inconsistency in coverage of NETs and their impacts highlights this key knowledge gap; this analysis will help to frame these developments in the modelling community.

For BECCS, research and development is required to deliver high efficiency energy conversion and distribution processes for the lowest impact CCS, and the cost of infrastructure to transport CO<sub>2</sub> from BECCS production areas to storage locations needs to be further evaluated. To

this end, early deployment of CCS would enhance understanding of the risks and possible improvements of the technology. Integrated pilot-plants need to be built (storing ~1 MtCO<sub>2</sub> per year) to examine how combined BECCS functions<sup>93</sup>; the capital cost of 5-10 full-size demonstrations of BECCS/CCS would require investment of approximately \$5-10 billion<sup>93</sup>. There is also a need to develop socio-economic governance systems for all NETs, to provide incentives to fund this research and development, and implementation of infrastructure in the most sustainable manner, to limit adverse impacts in the transition to low carbon energy systems, and to manage the risks associated with CCS (leakage, seismic action, environmental impacts)<sup>94</sup>. Priorities include investment in renewable and low carbon technologies, efficiency, and integration of energy systems (to make the most of waste heat, excess electrons from PV and wind, to close the carbon cycle of fossil sources by capture and reuse of CO<sub>2</sub> by catalysis), and the realisation of environmental co-benefits. In the meantime, emission reductions must continue to be the central goal for addressing climate change.

Addressing climate change remains a fundamental challenge for humanity, but there are risks associated with relying heavily on any technology that has adverse impacts on other aspects of regional or planetary sustainability to achieve our climate goals. Though deep and rapid decarbonisation may yet allow us to meet the <2°C climate goal through emissions reduction alone<sup>8</sup>, this window of opportunity is rapidly closing<sup>8,95</sup>, so there is likely to be some need for NETs in the future<sup>11,42</sup>. Our analysis indicates that there are numerous resource implications associated with the widespread implementation of NETs that vary between technologies and that need to be satisfactorily addressed before NETs could play a significant role in achieving climate change goals. Though some NETs could offer environmental co-benefits (e.g. improved soil carbon storage<sup>29</sup>), a heavy reliance on NETs in the future, if used as a means to allow continued use of fossil fuels in the present, is extremely risky since our ability to stabilise the climate at <2°C declines as cumulative emissions increase<sup>8,36,95</sup>. A failure of NETs to deliver expected mitigation in the future, due to any combination of biophysical and economic limits examined here, leaves us with no “Plan B”<sup>48</sup>. Since this study shows that there is no NET, or combination of NETs, available now that could be



implemented to meet the <2°C target without significant impact on either land, energy, water, nutrient, albedo or cost, “Plan A” must be to reduce GHG emissions aggressively now.

## References

- 1) Le Quéré, C. *et al.* The global carbon budget 1959-2011. *Earth System Science Data* **5**, 165-185 (2013).
- 2) Peters, G. P. *et al.* The challenge to keep global warming below 2°C. *Nature Clim. Change* **3**, 4-6 (2013). **Short article outlining the enormous challenge of meeting a 2°C climate stabilization target.**
- 3) Edenhofer, O. *et al.* (Eds) *Climate Change 2014: Mitigation of Climate Change. Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. (2014). **The latest IPCC Assessment Report on mitigation options available to stabilise climate.**
- 4) Tavoni, M. *et al.* Post-2020 climate agreements in the major economies assessed in the light of global models. *Nature Clim. Change* **5**, 119-126 (2015).
- 5) Krey, V., Luderer, G., Clarke, L. & Kriegler, E. Getting from here to there – energy technology transformation pathways in the EMF27 scenarios. *Climatic Change* **123**, 369-382 (2014).
- 6) Edmonds, J. *et al.* Can radiative forcing be limited to 2.6 Wm<sup>-2</sup> without negative emissions from bioenergy and CO<sub>2</sub> capture and storage? *Climatic Change* **118**, 29-43 (2013).
- 7) van Vuuren, D. P. *et al.* The role of negative CO<sub>2</sub> emissions for reaching 2 °C - insights from integrated assessment modelling. *Climatic Change* **118**, 15-27 (2013).
- 8) Rogelj, J., McCollum, D. L., Reisinger, A., Meinshausen, M. & Riahi, K. Probabilistic cost estimates for climate change mitigation. *Nature* **493**, 79-83 (2013).
- 9) Clarke, L. *et al.* Assessing transformation pathways. In: *Climate Change 2014: Mitigation of Climate Change. Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (ed. Edenhofer, O. *et al.*). Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. (2014).
- 10) Riahi, K. *et al.* Locked into Copenhagen pledges - Implications of short-term emission targets for the cost and feasibility of long-term climate goals. *Technological Forecasting and Social Change Part A*, **90**, 8-23 (2015).
- 11) Gasser, T., Guivarch, G., Tachiiri, K., Jones, C. D. & Ciais, P. Negative emissions physically needed to keep global warming below 2°C *Nature Comm.* doi:10.1038/ncomms8958 (2015).
- 12) Obersteiner, M. *et al.* Managing climate risk. *Science* **294**, 786-787 (2001).
- 13) Creutzig F. *et al.* Bioenergy and climate change mitigation: an assessment. *Glob. Change Biol. Bioenergy* doi:10.1111/gcbb.12205 (2015).
- 14) Keith, D. Why capture CO<sub>2</sub> from the atmosphere. *Science* **325**, 1654-1655 (2009).
- 15) Socolow, R. *et al.* *Direct air capture of CO<sub>2</sub> with chemicals: A technology assessment for the APS Panel on Public Affairs*. American Physical Society. 92pp. (2011). **In depth assessment of direct air capture technologies.**
- 16) Schuiling, R. D. & Krijgsman, P. Enhanced weathering: an effective and cheap tool to sequester CO<sub>2</sub>. *Climatic Change* **74**, 349–354 (2006).
- 17) Rau, G. H., Knauss, K. G., Langer, W. H. & Caldeira, K. Reducing energy-related CO<sub>2</sub> emissions using accelerated weathering of limestone. *Energy* **32**, 1471-1477 (2007).

- 18) Köhler, P., Hartmann, J. & Wolf-Gladrow, D. A. Geoengineering potential of artificially enhanced silicate weathering of olivine. *Proc. Natl Acad. Sci. USA* **107**, 20228-20233 (2010).
- 19) Hartmann, J. & Kempe, S. What is the maximum potential for CO<sub>2</sub> sequestration by “stimulated” weathering on the global scale? *Naturwissenschaften* **95**, 1159-1164 (2008).
- 20) Kelemen, P. B. & Matter, J. M. In situ carbonation of peridotite for CO<sub>2</sub> storage. *Proc. Natl Acad. Sci. USA* **105**, 17295-17300 (2008).
- 21) Arora, V. K. & Montenegro, A. Small temperature benefits provided by realistic afforestation efforts. *Nat. Geosci.* **4**, 514-518, (2011).
- 22) Canadell, J. G. & Raupach, M. R. Managing forests for climate change mitigation. *Science* **320**, 1456-1457 (2008).
- 23) Jackson, R. B. *et al.* Protecting climate with forests. *Environ. Res. Lett.* **3**, 044006 (2008).
- 24) Sarmiento, J. L., Gruber, N., Brzezinski, M. A. & Dunne, J. P. High-latitude controls of thermocline nutrients and low latitude biological productivity. *Nature* **427**, 56-60 (2004).
- 25) Joos, F., Sarmiento, J. L. & Siegenthaler, U. Estimates of the effect of Southern Ocean iron fertilization on atmospheric CO<sub>2</sub> concentrations. *Nature* **349**, 772-775 (1991).
- 26) Kheshgi, H. S. Sequestering atmospheric carbon dioxide by increasing ocean alkalinity. *Energy* **20**, 915-922 (1995).
- 27) Smith, P. Soils and climate change. *Curr. Opin. Environ. Sust.* **4**, 539–544 (2012).
- 28) Powlson, D. S. *et al.* Limited potential of no-till agriculture for climate change mitigation. *Nature Clim. Change* **4**, 678-683 (2014).
- 29) Smith, P. *et al.* Greenhouse gas mitigation in agriculture. *Phil. Trans. R. Soc. B* **363**, 789-813 (2008).
- 30) Woolf, D., Amonette, J. E., Street-Perrott, A., Lehmann, J. & Joseph, S. Sustainable biochar to mitigate global climate change. *Nature Comm.* **1**, Article 56, doi:10.1038/ncomms1053 (2010).
- 31) Schiermeier, Q. Convention discourages ocean fertilization. *Nature* (online; 12 November 2007) doi:10.1038/news.2007.230 (2007).
- 32) Smith, P. *et al.* How much land based greenhouse gas mitigation can be achieved without compromising food security and environmental goals? *Glob. Change Biol.* **19**, 2285–2302 (2013).
- 33) Smith P. *et al.* Agriculture, forestry and other land use (AFOLU). In: *Climate Change 2014: Mitigation of Climate Change. Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (ed. Edenhofer, O. *et al.*). Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. (2014).
- 34) Smith P. Soil carbon sequestration and biochar as negative emission technologies. *Glob. Change Biol.* (in review) (2015).
- 35) Azar, C. *et al.* The feasibility of low CO<sub>2</sub> concentration targets and the role of bio-energy carbon-capture and storage. *Climatic Change* **100**, 195-202 (2010).
- 36) Kriegler, E. *et al.* What does the 2°C target imply for a global climate agreement in 2020? The LIMITS study on Durban Platform scenarios. *Clim. Change Econ.* **04**, 1340008. doi: 10.1142/S2010007813400083 (2013).
- 37) Strengers, B. J., Minnen, J. G. V. & Eickhout, B. The role of carbon plantations in mitigating climate change: potentials and costs. *Climatic Change* **88**, 343-366 (2008).
- 38) Wise, M. *et al.* Implications of limiting CO<sub>2</sub> concentrations for land use and energy. *Science* **324**, 1183–1186 (2009).
- 39) Reilly, J. *et al.* Using land to mitigate climate change: hitting the target, recognizing the trade-offs *Environ. Sci. Technol.* **46**, 5672-5679 (2012).
- 40) Humpenöder, F. *et al.* Investigating afforestation and bioenergy CCS as climate change mitigation strategies. *Environ. Res. Lett.* **9**, 064029 (13pp) (2014).

- 41) Chen, C. & Tavoni, M. Direct air capture of CO<sub>2</sub> and climate stabilization: a model based assessment. *Climatic Change* **118**, 59-72 (2013).
- 42) Fuss, S. *et al.* Betting on negative emissions. *Nature Clim. Change* **4**, 850–853 (2014).
- 43) Kriegler E. *et al.* Is atmospheric carbon dioxide removal a game changer for climate change mitigation? *Climatic Change* **118**, 45–57 (2013).
- 44) Sims R. *et al.* Transport. In: *Climate Change 2014: Mitigation of Climate Change. Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (ed. Edenhofer, O. *et al.*). Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA (2014).
- 45) Kriegler, E. *et al.* Making or breaking climate targets: The AMPERE study on staged accession scenarios for climate policy. *Technological Forecasting and Social Change, Part A* **90**, 24-44. doi: 10.1016/j.techfore.2013.09.021. (2015). **Study showing the impact of delay in implementation of mitigation on climate stabilisation over the course of the 21<sup>st</sup> Century.**
- 46) Boden, T. A., Marland, G. & Andres, R. J. Global, regional, and national fossil-fuel CO<sub>2</sub> emissions. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy. Oak Ridge, Tenn., U.S.A. (2015).
- 47) Krey, V. *et al.* Annex II: Metrics and Methodology, Section A.II.10: Scenario Data. In: *Climate Change 2014: Mitigation of Climate Change. Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (ed. Edenhofer, O. *et al.*). Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. (2014).
- 48) National Academy of Sciences *Climate Intervention: Carbon Dioxide Removal and Reliable Sequestration*. The National Academies Press, Washington DC, USA, 140pp, (2015). **In depth report on carbon dioxide removal (equivalent to negative emissions) technologies.**
- 49) Calvin, K. *et al.* A multi-model analysis of the regional and sectoral roles of bioenergy in near- and long-term CO<sub>2</sub> emissions reduction. *Climate Change Economics* **4**, 1340014 (32 pages). (2013).
- 50) Popp, A. *et al.* Land-use transition for bioenergy and climate stabilization: model comparison of drivers, impacts and interactions with other land use based mitigation options. *Climatic Change* **123**, 495-509 (2014).
- 51) Bonsch, M. *et al.* Trade-offs between land and water requirements for large-scale bioenergy production. *Glob. Change Biol. Bioenergy* doi: 10.1111/gcbb.12226 (2014).
- 52) Wieder, W. R., Cleveland, C. C., Smith, W. K. & Todd-Brown, K. Future productivity and carbon storage limited by terrestrial nutrient availability. *Nat. Geosci.* **8**, 441-444 (2015).
- 53) Smith, L. J. & Torn, M. S. Ecological limits to terrestrial biological carbon dioxide removal. *Climatic Change* **118**, 89-103 (2013). **Study examining some ecological limits to land based negative emission technologies.**
- 54) Scott, V., Haszeldine, R. S., Tett, S. F. B. & Oschlies, A. Fossil fuels in a trillion tonne world. *Nature Clim. Change* **5**, 419-423 (2015).
- 55) Benson, S.M. *et al.* (2012), Carbon capture and Storage, Chapter 13 in *Global Energy Assessment Toward a Sustainable Future*, Cambridge University Press. (Online ISBN:9780511793677; Hardback ISBN:9781107005198; Book DOI: <http://dx.doi.org/10.1017/CBO9780511793677>).
- 56) Zhao, K. & Jackson, R. B. Biophysical forcings of land-use changes from potential forestry activities in North America. *Ecol. Monogr.* **84**, 329-353 (2014).
- 57) Zhuang, Q., Qin, Z. & Chen, M. Biofuel, land and water: maize, switchgrass or Miscanthus? *Environ. Res. Lett.* **8**, 015020 (6pp) (2013).
- 58) Edenhofer, O. *et al.* (eds) *IPCC Special Report on Renewable Energy Sources and Climate Change Mitigation*. Cambridge University Press, New York, NY, USA (2011).

- 59) Renforth, P. The potential of enhanced weathering in the UK. *Int. J. Greenh. Gas Con.* **10**, 229–243 (2012).
- 60) Christian, D., Riche, A. B. & Yales, N. E. Growth, yield and mineral content of *Miscanthus × giganteus* grown as a biofuel for 14 successive harvests. *Ind. Crop Prod.* **28**, 320–327 (2008).
- 61) St Clair, S., Hiller, J. & Smith, P. Estimating the pre-harvest greenhouse gas costs of energy crop production. *Biomass Bioenerg.* **32**, 442–452 (2008).
- 62) Swann, A. L., Fung, I. Y., Levis, S., Bonan, G. B. & Doney, S. C. Changes in Arctic vegetation amplify high-latitude warming through the greenhouse effect. *Proc. Natl Acad. Sci. USA* **107**, 1295–1300 (2010).
- 63) Ma, D., Notaro, M., Liu, Z.Y., Chen, G.S. & Liu, Y. Q. Simulated impacts of afforestation in East China monsoon region as modulated by ocean variability. *Clim. Dynam.* **41**, 2439–2450 (2013).
- 64) Betts, A.K. & Ball, J.H. Albedo over the boreal forest. *J. Geophys. Res.* **102 (D24)**, 28901–28910. doi:10.1029/96JD03876 (1997).
- 65) Betts R.A. Offset of the potential carbon sink from boreal forestation by decreases in surface albedo. *Nature* **408**, 187–190 (2007).
- 66) Betts R.A., Falloon, P. D., Goldewijk, K. K. & Ramankutty, N. Biogeophysical effects of land use on climate: Model simulations of radiative forcing and large-scale temperature change. *Agr. Forest Meteorol.* **142**, 216–233 (2007).
- 67) Schaeffer, M. *et al.* CO<sub>2</sub> and albedo climate impacts of extratropical carbon and biomass plantations. *Global Biogeochem. Cy.* **20**, GB2020, doi:10.1029/2005GB002581 (2006).
- 68) Jones, A. D. *et al.* Greenhouse gas policy influences climate via direct effects of land-use change. *J. Climate* **26**, 3657–3670 (2013).
- 69) Mazzotti, M., Bociocchi, R., Desmond, M. J. & Socolow, R. Direct air capture of CO<sub>2</sub> with chemicals: optimization of a two-loop hydroxide carbonate system using a countercurrent airliquid contactor. *Climatic Change* **118**, 119–135 (2013).
- 70) Klein, D. *et al.* The value of bioenergy in low stabilization scenarios: an assessment using REMIND-MAGPIE. *Climatic Change* **123**, 705–718 (2014).
- 71) Lenton, T. M. The global potential for carbon dioxide removal. In: *Geoengineering of the Climate System* (ed. Harrison, R. M. & Hester, R. E.) Royal Society of Chemistry, London, UK (2014).
- 72) Haberl, H. *et al.* Quantifying and mapping the human appropriation of net primary production in earth's terrestrial ecosystems. *Proc. Natl Acad. Sci. USA* **104**, 12942–12947 (2007).
- 73) Schueler, V., Weddige, U., Beringer, T., Gamba, L. & Lamers, P. Global biomass potentials under sustainability restrictions defined by the European Renewable Energy Directive 2009/28/EC. *Glob. Change Biol. Bioenergy* **5**, 652–663 (2013).
- 74) FAOSTAT <http://faostat3.fao.org/home/E> (accessed 25 June 2015) (2015).
- 75) Canadell, J.G., Schulze, E.-D. Global potential of biospheric carbon management for climate mitigation. *Nature Comm.* **5**, 5282: 1–12. doi: 10.1038/ncomms6282 (2014).
- 76) Smith, P. *et al.* Competition for land. *Phil. Trans. R. Soc. B* **365**, 2941–2957 (2010).
- 77) Thompson, B. & Cohen, M. J. *The impact of climate change and bioenergy on nutrition*. Springer, Rome, Italy (2012).
- 78) Valentine, J., Clifton-Brown, J., Hastings, A., Robson, P., Allison G. & Smith, P. Food vs. fuel: the use of land for lignocellulosic 'next generation' energy crops that minimise competition with primary food production. *Glob. Change Biol. Bioenergy* **4**, 1–19 (2012).
- 79) Bustamante, M. *et al.* Co-benefits, trade-offs, barriers and policies for greenhouse gas mitigation in the Agriculture, Forestry and Other Land Use (AFOLU) sector. *Glob. Change Biol.* **20**, 3270–3290 (2014).
- 80) Powell, T. W. R. & Lenton, T. M. Future carbon dioxide removal via biomass energy constrained by agricultural efficiency and dietary trends. *Energy Environ. Sci.* **5**, 8116–8133 (2012).

- 81) Smith, P. Delivering food security without increasing pressure on land. *Global Food Security* **2**, 18-23 (2013).
- 82) Bajželj, B. *et al.* The importance of food demand management for climate mitigation. *Nature Clim. Change* **4**, 924–929 (2014).
- 83) Vörösmarty, C. J. Global water resources: vulnerability from climate change and population growth. *Science* **289**, 284-288 (2000).
- 84) Stolaroff, J. K., Keith, D. W. & Lowry, G. V. Carbon dioxide capture from atmospheric air using sodium hydroxide spray. *Environ. Sci. Technol.* **42**, 2728–2735 (2008).
- 85) Oki, T. & Kanae, S. Global hydrological cycles and world water resources. *Science* **313**, 1068-1072 (2006).
- 86) Postel, S. L., Daily, G. C. & Ehrlich, P. R. Human appropriation of renewable freshwater. *Science* **271**, 785-788 (1996).
- 87) Rockstrom, J. *et al.* The unfolding water drama in the Anthropocene: towards a resilience-based perspective on water for global sustainability. *Ecohydrol.* **7**, 1249-1261 (2014).
- 88) Creutzig, F. *et al.* Reconciling top-down and bottom-up modelling on future bioenergy deployment. *Nature Clim. Change* **2**, 320-327 (2012).
- 89) Cowie, A. L., Smith, P. & Johnson, D. Does soil carbon loss in biomass production systems negate the greenhouse benefits of bioenergy? *Mitigation Adapt. Strateg. Glob. Chang.* **11**, 979-1002 (2006).
- 90) McCollum, D. *et al.* Energy investments under climate policy: a comparison of global models. *Climate Change Economics* **4**, 1340010, doi: 10.1142/S2010007813400101 (2013).
- 91) Creutzig, F. Economic and ecological views on climate change mitigation with bioenergy and negative emissions. *Glob. Change Biol. Bioenergy* doi:10.1111/gcbb.12235 (2015).
- 92) Kato, E. & Yamagata, Y. BECCS capability of dedicated bioenergy crops under a future land-use scenario targeting net negative carbon emissions. *Earth's Future* **2**, 421–439 (2014).
- 93) Scott, V., Gilfillan, S., Markusson, N., Chalmers, H., Haszeldine, R. S. Last chance for carbon capture and storage. *Nature Clim. Change* **3**, 105-111 (2012).
- 94) Blackford, J. *et al.* Detection and impacts of leakage from sub-seafloor deep geological carbon dioxide storage. *Nature Clim. Change* **4**, 1011-1016 (2014).
- 95) Luderer, G. *et al.* Economic mitigation challenges: how further delay closes the door for achieving climate targets. *Environ. Res. Lett.* **8**, 034033 (2013). **Study showing the urgency of climate mitigation action.**

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### Author contributions

**PS** led the writing of the paper, with contributions from **all authors** in the inception of the study and in writing the drafts. **PS** led the analysis with significant contributions from **SJD, FC, SF, JM, BG, RBJ, AC, EKr, DM** and **DVV**. Figures were conceptualised and produced by **SJD, JR, PC, SF, PS, GP, RA** and **JM**.

### Competing financial interests

MO was given a share in Biorecro, a company that cooperates with BECCS projects globally, honouring his pioneering work on BECCS. The other authors have no competing financial interests to declare.

### Figure legends

**Figure 1.** Schematic representation of carbon flows among atmospheric, land, ocean and geological reservoirs. Climate change results from addition of geological carbon to the atmosphere *via* combustion or other processing of fossil fuels for energy (**A**). Bioenergy seeks to avoid the net addition of carbon to the atmosphere by instead utilizing biomass energy at a rate that matches the uptake of carbon by re-growing bioenergy feedstocks (**B**). Carbon capture and storage (CCS) technologies intervene to capture most of the potential carbon emissions from fossil fuels, and return them to a geological (or possibly ocean) reservoir (**C**). NETs remove carbon from the atmosphere, either *via* biological uptake (**G** and **H**), or, uptake *via* biological or industrial processes with CCS (**D** and **E**), or enhanced weathering of minerals (**F**). Any atmospheric perturbation will lead to re-distribution between the other reservoirs, but these homeostatic processes are not shown. Note that there are significant differences in the materials and energy requirements for each process to remove or avoid a unit mass of carbon from (or to) the atmosphere.

**Figure 2.** Scenarios including NETs for each of the scenario categories, corresponding to the ranges and median values shown in Table S3. Scenarios with no technology constraints (i.e. including NETs) from the AMPERE<sup>10,45</sup> and LIMITS<sup>36</sup> modelling comparison exercises are shown in colours (as per key for different CO<sub>2</sub>-eq. concentration ranges), with all other scenarios from the IPCC AR5 database

shown in grey. See Table S3 caption for explanation of representation of gross positive and gross negative emissions. Net land use change fluxes are included (note 1997 fluctuation attributable to Indonesian peat fires). Sources: CDIAC<sup>46</sup>, IPCC AR5 scenario database (available at: <https://secure.iiasa.ac.at/web-apps/ene/AR5DB/>)<sup>47</sup>, Global Carbon Project.

**Figure 3.** Negative emissions technologies have different land (A), water (B), and nutrient (C) requirements, have different geophysical impacts on climate (e.g. albedo, D), generate or require different amounts of energy (E), and entail different capital and operating costs (F). For instance, carbon dioxide removal (CDR) technologies such as DAC and EW of silicate rock tend to require much less land and water than strategies that depend on photosynthesis to reduce atmospheric carbon (A and B), but the CDR technologies demand substantial energy and economic investment per unit of negative emissions (E and F). Among BECCS options, forest feedstocks tend to require less nitrogen than purpose-grown crops (C), but present greater risk of unwanted changes in albedo (D), and with less energy generated (E). AR omitted from B, E and F to avoid confusion with forest BECCS (where CCS component is included). See Methods Table S1 for sources.

**Figure 4.** Schematic representation of the aggregate impacts of NETs on land, energy, water, and relative investment needs for levels of implementation equivalent to BECCS (3.3 GtC/yr in 2100) in scenarios consistent with a 2°C target (or mean, maximum attainable where that level of negative emissions cannot be reached). Water requirement shown as water drops with quantities in km<sup>3</sup>/yr. All values are for 2100 except relative costs which are for 2050 (see Methods).

**Table 1.** Global impacts of NETs for the average needed global C removals per year in 2100 in 2°C-consistent scenarios (430-480ppm scenario category; Table S3). The NETs with lower maximum potential than the BECCS emission requirement of 3.3 GtCeq./year in 2100 are coloured grey and their mean (maximum) potential is given along with their impacts (see Methods). Wide ranges exist for most impacts but for simplicity and to allow comparison between NETs (sign and order of magnitude), mean values are presented. See Methods and text for full details.

NET	Global C removal (GtCeq./yr in 2100)	Mean (max), land requirement (Mha in 2100)	Estimated energy requirement (EJ/yr in 2100)	Mean (max), water requirement (km <sup>3</sup> /yr in 2100)	Nutrient impact (ktN/yr in 2100)	Albedo impact in 2100	Investment needs (BECCS for electricity / BECCS for biofuel; B\$/yr in 2050)
BECCS	3.3	380-700	-170	720	Variable	Variable	138 / 123
DAC	3.3	Very low (unless solar PV used for energy)	156	10-300	None	None	>> BECCS
EW	0.2 (1.0)	2 (10)	46	0.3 (1.5)	None	None	>BECCS
AR	1.1 (3.3)	320 (970)	Very low	370 (1040)	2.2 (16.8)	Negative; or reduced GHG benefit where not negative	<<BECCS



## Methods

We describe the sources of information used in the analysis below; knowledge levels and uncertainties across the different NETs, and in terms of demand on land, water, nutrients, albedo, energy and costs differ greatly, but are sufficient to inform this comparative analysis. We have used all literature able to inform the analysis, irrespective of methodological approaches taken (e.g. attributional and consequential analyses are used as appropriate). Where possible, we have calculated impacts relative to counterfactual estimates (e.g. evaporative loss for BECCS and AR). Table S1 presents a summary of the data sources used to estimate per-t-C and global impact ranges, and provides a qualitative assessment of the relative confidence / uncertainty in the estimated ranges.

[Table S1 here]

The net effect of NETs on radiative forcing depends on equilibrium effects in markets, and biophysical dynamic effects, subject only to general, or at least partial equilibrium models. Such models would *inter alia* specify changes in deployment of other technologies, as a result of NET deployment, and model the ensuing effects on water, land, nutrients, etc.<sup>13,88,96</sup>. The results of our study can be used as input values to global energy-system, climate and integrated assessment models that enable the investigation of dynamic effects.

### ***Impacts on land / GHG emissions, water, energy, nutrients, albedo and cost on a per-t-C removed from the atmosphere basis***

Estimates of impacts on land / GHG emissions, water, energy, nutrients, albedo and cost on a per-t-Ceq. removed from the atmosphere basis (as used shown in Figure 3) were derived as follows.

*Land / GHG emissions:* Annual net removals of carbon from the atmosphere per unit area (t Ceq./ha/yr) were estimated as follows: DAC - Quantity of C removed per year by a reference DAC facility of 1 Mt CO<sub>2</sub> = 0.27 Mt C covering an area (including spacing and on-site compressor and regenerator)<sup>15</sup> of 150 ha; AR – assumes 500 tCO<sub>2</sub>/ha = 136 tC/ha from forest regrowth over 40 years

to maturity (assumes linear uptake), so mean annual accrual rate is 3.4 tC/ha/yr – for longer lasting AR, annual accrual rates drop (i.e. annual accrual rate over 80 years would be half of that over 40 years); For BE crops, negative emissions exclude fossil fuel displacement and are assumed to be mean of the range of carbon in biomass available for capture. Dedicated BE crop values are from Table S2 (mean of range for Miscanthus); Marginal crops - values from Table S2 (mean of the range for Sorghum); Crop residues - values from Table S2 (mean of the range for crop residues); Coppice - values from Table S2 (willow/poplar SRC); Pine - assumed same as annual increment for AR (see above); Tropical forests - tropical forests in humid areas assumed to have ~3x higher C accumulation than temperate forests following<sup>97</sup>; Boreal forests - boreal coniferous forests are assumed to have about half the carbon accumulation of temperate coniferous forests following<sup>97</sup>; EW of olivine value is the maximum value from<sup>19</sup>. Crop and forest BECCS sources are from<sup>3,64,56,98-101</sup>. Land use intensity (the area of land to produce one tCeq. of negative emissions) is plotted in Figure 3A.

[Table S2 here]

In every case, the land use intensity increases as lower fertility land is used. Associated with the land-based NETs are carbon stock changes and greenhouse gas (GHG) emissions, which need to be accounted for when calculating the net GHG balance. Even partial removal (30-40%) of crop and forest residue from land increases soil erosion hazard, soil carbon and nutrient losses<sup>102</sup>, and similar impacts occur with forest residue removal<sup>103</sup>, so care must be taken over residue removal rates, which can reduce the quantity of residue available for NETs considerably. Using organic waste (e.g. animal manure or residues from food processing that are otherwise used as soil amendment) presents similar constraints, although some energy conversion processes make it possible to return part of this organic matter to soils (e.g. anaerobic digestion<sup>104</sup>). Overall, the soil C sequestration forgone and nutrients exported by removing residues means that residue use as a feedstock is not GHG neutral (as the counterfactual would result in higher soil carbon stocks), but life-cycle GHG emissions are lower than for some BE crops. Edible feedstocks (i.e. which are used to produce most currently available biofuels) entail large N<sub>2</sub>O emissions related to inputs of fertilizer N, accounting for up to half of the life-cycle GHG emissions of the end-products<sup>105</sup>, in addition to their indirect emissions through land-

use change effects. Lignocellulosic biomass crops (e.g. miscanthus, switchgrass, sweet sorghum) emit up to 10 times less N<sub>2</sub>O than food crops due to lower fertiliser N requirement, and have the potential to increase soil organic carbon if perennial<sup>106</sup>, possibly resulting in a GHG sink were perennials to substitute for arable crops and little fertilizer were used. A major unknown is the global land-use change effects of purpose-grown BE crops given the possibility of land use change in one place driving conversion of land elsewhere<sup>50,107-109</sup>. Growing BE crops on marginal land could reduce indirect land-use change and associated GHG emissions, but productivity on this land is likely to be relatively low, leading to lower uptake / higher GHG emissions per MJ, compared to more productive land<sup>110-111</sup>. Table S2 shows the carbon and GHG emissions / removals associated with a range of energy crops and forest types, and the net negative emissions obtained including fossil-fuel offsets.

*Water:* Annual water use per-t-C removed from the atmosphere (m<sup>3</sup>/tCeq./yr) values were estimated as follows: DAC – minimum, maximum and average water use values are from estimates of evaporative loss in amine DAC units given in<sup>84</sup>; AR – provides total interception and transpiration water loss for broadleaves (low value of range) and conifers (high value of range)<sup>112</sup> calculated assuming annual rainfall of 1000mm and annual increments given in the land / GHG emissions panel. All water use estimates subtract the water use under previous land use to provide a net change in water use. For all of the following BE sources, annual water use for growing the crop was added to water for cooling the plant and CCS technology. The CCS component of BECCS is also relatively water intensive. Extra water is needed for the scrubbers that remove CO<sub>2</sub> from the air compared to a power plant without CCS. Additional water is also typically needed for the energy penalty of the carbon-capture system, in particular the energy needed to power the CCS. This parasitic load reduces a plant's capacity by ~20-25%<sup>113</sup>. Another analysis<sup>114</sup> estimated that a comprehensive implementation of CCS in 2030 could raise freshwater withdrawals by 2–3% and consumption by as much as 52–55% in the United States. Though there is a range of potential water use characteristics for CCS plants<sup>115</sup>, the CCS component of BECCS is assumed to have the same water requirements as a coal CCS plant<sup>15</sup>. Annual water use for plant growth was calculated as follows: BE crops – annual water use assumed as per broadleaved trees above; Marginal crops - annual water use value for sorghum from<sup>116</sup>; Crop

residues - annual water use value for wheat from<sup>116</sup>; Coppice – annual water use assumed the same as broadleaved forest; Pine – annual water use assumed the same as coniferous forest; Tropical forest - annual water use assumed the same as broadleaved forest; Boreal forest - annual water use assumed the same as coniferous forest – all forests are assumed to be unirrigated. For EW of olivine, one molecule of water is required for each molecule of CO<sub>2</sub> removed, so each tCeq. would require 1.5 m<sup>3</sup> water. Expressing additional water use as a proportion of runoff in a region would give a more accurate picture of the threat to water resources at a given location, but without a spatially disaggregated analysis this is not feasible, so global assessments relative to total freshwater use are presented.

*Energy:* Energy production or energy input requirement per-t-C removed from the atmosphere (GJ/tCeq./yr) were estimated as follows: DAC – minimum of the range is the combined minimum thermodynamic energy for capture plus minimum theoretical compression energy, and maximum of the range assumes 40% conversion efficiency for heat and electricity with all values from page 40 of<sup>15</sup>; EW minimum energy requirements are calculated from open-pit mining energy consumption<sup>117</sup>, and olivine CO<sub>2</sub> capture potential<sup>116</sup>; with total energy requirement estimates from<sup>57</sup>; AR requires low energy input (for site preparation only) and has no energy output. Energy end uses for BECCS vary, but here we calculate for power generation (using the median value reported in<sup>3</sup>, i.e. conversion technologies representative of the 2000-2010 time period worldwide), where the carbon can be captured for storage. For all of the following BE sources, energy use for CCS technology for a BECCS plant has the same energy requirements as a coal CCS plant from page 25 of<sup>15</sup>, and CCS energy use is subtracted from the energy generated from combustion of the BE feedstock (GJ/tC), with assumed energy penalties of 20-25% (Table S2<sup>118</sup>). BE crops – ranges for oil given by values for Brazilian Soybean (low) and Asian oil palm (high), ranges for starch/sugar ethanol given by European wheat (low) and Brazilian sugarcane (high), ranges for lignocellulose given by North American switchgrass (low) and European Miscanthus (high) – all from<sup>58</sup>; Marginal crops – range for oil given by Indian Jatropha (low) and Jatropha from Thailand (high) from<sup>58</sup>; Crop residues - range for lignocellulose given by Sorghum stover (low) and corn stover (high) from<sup>58</sup>; Coppice – range given

by the range for coppice for Europe from<sup>58</sup>; Pine – annual increment for pine is ~half of increment for coppice so energy output is assumed to be half of coppice; Tropical forest – has ~3x higher C accumulation than temperate forests following Table 3A.5 of<sup>97</sup> so energy output assumed to be 3x pine; Boreal forest - assumed to have about half the carbon accumulation of temperate coniferous forests following Table 3A.5 of<sup>97</sup> so energy output assumed to be half that of pine.

*Nutrients:* Nutrient content (here represented by nitrogen content: kg N kg C<sup>-1</sup>) is not applicable to DAC or EW technologies. For forests and energy crops, all values are from<sup>88,101</sup>. Most modern lignocellulosic energy crops require no annual application of N fertiliser (though a small amount is sometimes used in the establishment phase<sup>60</sup> – but usually much less than applied to cropland and pastureland that is replaced). Further, the ratio of N<sub>2</sub>O emissions to fertilizer N inputs is much lower for perennial BE crops than it is for cropland and grassland<sup>106</sup>. A recent study on growing wheat after growing unfertilized miscanthus for 20 years showed very little depletion of soil N<sup>119</sup>.

*Albedo:* DAC and EW assumed to have no impact on albedo. All values for albedo change (unit-less) are for surface albedo. These values for forests and BE feedstocks are from<sup>56,64</sup> and assume change from grassland. Values for broadleaves (aspen) used for BE crops, coppice and tropical forest, and values for conifers are used for pine and boreal forests. The range for AR uses values for broadleaves (high) and conifers (low). Values including and excluding snow cover are presented.

*Costs:* Costs per unit of carbon removed from the atmosphere (\$ tC<sup>-1</sup>) of DAC are from page 14 of<sup>45</sup>. Costs of enhanced weathering of olivine come from<sup>16,57</sup>. Costs for BECCS are the range from 6 IAMs<sup>49</sup> and the range reflects the type and energy end-use of BECCS and regional variability. These \$/tC values are shown in Figure 3 but are not used for upscaling to total global costs (see below).

### ***Implementation of NETs***

Levels of implementation of NETs consistent with a <2°C target (i.e. with concentration levels of 430–480 ppm CO<sub>2</sub>-eq. in 2100) were assumed; for BECCS this is 3.3 GtCeq./yr in 2100 (Table S3<sup>10,36,45</sup>). For DAC, though the maximum level of deployment could yield ~10 GtCeq./yr removals<sup>41</sup>

in 2100, for comparison with BECCS we assume the level of implementation of DAC that also delivers 3.3 GtCeq./yr negative emissions in 2100. For other NETs which are not able to meet the same level of removals, we use values compiled from an analysis of the recent literature to give mean and maximum implementation levels. The area under AR is not given in the AMPERE<sup>10,45</sup> and LIMITS<sup>36</sup> studies, so AR impacts were estimated for areas calculated using the mean AR accrual rate of 3.4 tCeq./ha/yr (Fig. 3A), at implementation levels in 2100 estimated to give removals in 2100 of around 1.1 GtCeq./yr, with a maximum value of 3.3 GtCeq./yr (from<sup>6,7,71</sup>). EW estimates are mean and maximum carbon removal from ocean liming and addition of crushed olivine to the ocean, and “other” is for EW by soil loading<sup>71</sup> giving mean removals of 0.2 Pg C yr<sup>-1</sup> by 2100 and maximum values of 1 Pg yr<sup>-1</sup>.

[Table S3 here]

### ***Bottom-up estimation of global impacts and limits to supply of NETs***

Bottom-up estimates of global impacts and limits to supply of NETs were estimated by multiplying the per-t-C impact estimates of impact (see Figure 3) by the total levels of implementation of each NET expressed as GtCeq./yr or Mha/yr in 2100, described above. Since costs cannot be scaled using per-t-C impacts, investment needs were used instead as described in *Investment needs* below.

*Investment needs:* Investments into BECCS technologies provide an additional indicator for assessing the scale and speed of BECCS deployment over the next several decades. Table S4 summarizes investment estimates<sup>90</sup> from six global integrated assessment models that assessed 2°C scenarios within the context of the LIMITS model inter-comparison (one of the studies contributing to the study summarised in Table S4). Owing to unique assumptions in the models, there are considerable differences in capital requirements for biomass electricity generation with CCS and biofuels production with CCS by 2030 and 2050. In fact, some models prefer a single route to negative emissions while completely foregoing another. On average across the models, some \$36.2 and \$29.4 billion/yr worth of investment is seen as optimal by 2030 for scaling up biomass electricity and

biofuels production technologies worldwide. By 2050, these investment levels grow to \$138.3 and \$122.6 billion/yr, respectively. In the near term (2030), BECCS investments appear to be split fairly evenly between today's industrialized and developing countries, whereas in the mid-term (2050) the IAMs indicate that the bulk of the investment dollars will likely need to flow to the developing world. That does not imply, however, that developing countries will be responsible for bearing the full costs of these negative emissions efforts. In particular, by mid-century, China, the United States and the countries of Latin America and Southeast Asia are projected to invest heavily in BECCS technologies.

[Table S4 here]

## References

- 96) Plevin, R.J., Delucchi, M., & Creutzig, F. Using attributional life cycle assessment to estimate climate-change mitigation benefits misleads policy makers. *J. Indust. Ecol.* **18**, 73-83 (2014).
- 97) IPCC *Good Practice Guidance for Land Use, Land-Use Change and Forestry*. Available at: <http://www.ipcc-nggip.iges.or.jp/public/gpglulucf/gpglulucf.html>. (Accessed 25th June 2015) (2003).
- 98) Macknick, J. *et al.* Operational water consumption and withdrawal factors for electricity generating technologies: a review of existing literature. *Environ. Res. Lett.* **7**, 045802 (2012).
- 99) Lackner, K.S. (pers. comm.)
- 100) Phyllis2 Database. Available at: <https://www.ecn.nl/phyllis2/> (Accessed 25th June 2015) (2015).
- 101) Gabrielle, B. *et al.* Paving the way for sustainable bioenergy in Europe: technological options and research avenues for large-scale biomass feedstock supply. *Renew. Sust. Energ. Rev.* **33**, 11-25 (2014).
- 102) Lal, R. World crop residues production and implications of its use as a biofuel. *Environ. Int.* **31**, 575-584 (2005).
- 103) Repo, A., Böttcher, H., Kindermann, G., & Liski, J. *et al.* Sustainability of forest bioenergy in Europe: land-use-related carbon dioxide emissions of forest harvest residues. *Glob. Change Biol. Bioenergy* **7**, 877-887 (2014).
- 104) Smith, J. *et al.* What is the potential for biogas digesters to improve soil fertility and crop production in Sub-Saharan Africa? *Biomass Bioenerg.* **70**, 58-72 (2014).
- 105) Smeets, E. M. W., Bouwman, L. F., Stehfest, E., van Vuuren, D. P. & Postuma, A. Contribution of N<sub>2</sub>O to the greenhouse gas balance of first-generation biofuels. *Glob. Change Biol.* **15**, 1-23 (2009).
- 106) Don, A. *et al.* Land-use change to bioenergy production in Europe: implications for greenhouse gas balance and soil carbon. *Glob. Change Biol. Bioenergy* **4**, 372-391 (2012).

- 107) Calvin, K. *et al.* Trade-offs of different land and bioenergy policies on the path to achieving climate targets. *Climatic Change* **123**, 691-704 (2014).
- 108) Searchinger, T. *et al.* Use of US croplands for biofuels increases greenhouse gases through emissions from land-use change. *Science* **319**, 1238–1240 (2008).
- 109) Havlik, P. *et al.* Global land-use implications of first and second generation biofuel targets. *Energy Policy* **39**, 5690–5702 (2011).
- 110) Field, C. B., Campbell, J. E. & Lobell, D. B. Biomass energy: the scale of the potential resource. *Trends Ecol. Evol.* **23**, 65-72 (2008).
- 111) Vuichard, N., Ciais, P. & Wolf, A. Soil carbon sequestration or biofuel production: new land-use opportunities for mitigating climate over abandoned Soviet farmlands. *Environ. Sci. Tech.* **43**, 8678-8683 (2009).
- 112) Forestry Commission. *Water use by trees*. Information Note, Forestry Commission. Available at: [http://www.forestry.gov.uk/pdf/FCIN065.pdf/\\$FILE/FCIN065.pdf](http://www.forestry.gov.uk/pdf/FCIN065.pdf/$FILE/FCIN065.pdf) (Accessed 25<sup>th</sup> June 2015) (2005).
- 113) Rochelle, G. T. Amine scrubbing for CO<sub>2</sub> capture. *Science* **325**, 1652–1654 (2009).
- 114) National Energy Technology Laboratory *Estimating freshwater needs to meet future thermoelectric generation requirements*. Pittsburgh, PA, 2009, DOE/NETL-400/2009/1339 (2009).
- 115) Chandel, M. K., Pratson, L. F. & Jackson, R. B. The potential impacts of climate-change policy on fresh water use in thermoelectric power generation. *Energy Policy* **39**, 6234–6242 (2011).
- 116) WWI Available at: <http://vitalsigns.worldwatch.org/node/180>. (Accessed 25th June 2015) (2013).
- 117) CIPEC Benchmarking the energy consumption of Canadian open-pit mines. Available at: <http://www.nrcan.gc.ca/sites/oe.nrcan.gc.ca/files/pdf/publications/industrial/mining/open-pit/Open-Pit-Mines-1939B-Eng.pdf> (Accessed 25<sup>th</sup> June 2015) (2005).
- 118) Gough, C. & Upham, P. *Biomass energy with carbon capture and storage (BECCS): a review*. Tyndall Centre for Climate Change Research, Working Paper 147. 40pp. Available at: <http://www.tyndall.ac.uk/sites/default/files/twp147.pdf>. (Accessed 25th June 2015) (2010).
- 119) Dufossé, K., Drewer, J., Gabrielle, B. & Drouet, J.-L. Effects of a 20-year old Miscanthus × giganteus stand and its removal on soil characteristics and greenhouse gas emissions. *Biomass Bioenergy* **69**, 198-210 (2014).



**Table S1.** Data sources used to estimate per-t-C and global impact ranges of NETs. OC = own calculations; n/a = not applicable; Numbers refer to references from which data were drawn or used in own calculations; Colours are qualitative assessment of confidence / uncertainty in the estimated ranges where green = relatively high confidence / low uncertainty in the estimated range, red = relatively low confidence / high uncertainty in the estimated range, and orange = medium confidence / uncertainty in estimated range.

NET	Per-t-C							Global estimates						
	Negative emission	Land intensity	Energy	Water	Nutrients	Albedo	Cost	Negative emission	Land use	Energy	Water	Nutrients	Albedo	Cost
BECCS	OC, 58	OC, 58	OC, 15, 58, 118	OC, 15, 56, 98, 113, 115, 116	OC, 60, 88, 100, 101, 106	64	49, 57, 58, 70	10, 36, 45	OC, 40, 42, 50, 53, 73	OC, 10, 36, 45, 73, 88, 91	OC, 51, 53	OC, 53	OC, 64	90
DAC	15	15	15	15, 84	n/a	n/a	15, 69	OC, 15, 41	OC	OC, 15	15, 84	n/a	n/a	OC – qualitative
EW	19	19	16, 59, 117,	59	n/a	n/a	16, 59	71	OC	OC, 59	59	n/a	n/a	OC – qualitative
AR	OC, 97	OC, 58, 98	OC, 97	OC, 56, 112	OC, 88, 100	64, 66, 67, 68	49	OC, 6, 7, 71	OC, 40, 42, 50, 53	n/a	OC, 40	OC, 52, 53	OC, 65	OC - qualitative

**Table S2.** Contribution of different energy crops and residue types to reducing GHG emissions though use as BE feedstock<sup>58,100,101,106</sup>. The column “Carbon in biomass available for capture” (shown in bold) is used in the calculation of negative emissions potential of BECCS.

Crop type	N <sub>2</sub> O emissions	Additional soil organic carbon (relative to annual food crops)	Additional below-ground biomass carbon	Temporary carbon storage in above-ground biomass	Carbon emission from indirect land-use change	Overall balance (as negative emissions)	<b>Carbon in biomass available for capture</b>	Total emission reduction including fossil fuel displacement and carbon capture	Total emission reduction including fossil fuel displacement and carbon capture (mean of range)
	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>	<i>t Ceq./ha/yr</i>
Miscanthus	0	0.68	0.35 - 0.46	0	0-0.29	0.75 – 1.15	<b>5.83-8.59</b>	7.54 – 15.68	11.61
Switchgrass	0	1.0	0.15 - 0.26	0	0-0.53	0.63 – 1.26	<b>3.16-4.60</b>	4.30 – 9.04	6.67
Willow / Poplar SRC	0	0.44	0.05 - 0.24	0-0.75	0-0.39	0.1 – 1.43	<b>4.67</b>	5.66 – 9.71	7.69
Eucalyptus	0	0.44	0.05 - 0.24	0-1.0	0-0.43	0.06 – 1.68	<b>4.17-11.53</b>	5.02 – 22.14	13.58
Annual crops (e.g. sorghum)	0.55	-0.19	0	0	0.36	-1.10 - -0.74	<b>4.60-11.96</b>	4.25 – 19.50	11.88
Residues (agriculture)	0.07	-0.19	0	0	0	-0.26	<b>1.66-1.78</b>	1.67 – 2.75	2.21
Residues (forestry)	0	-0.06	0	0	0	-0.05	<b>0.60-1.05</b>	0.65 – 1.80	1.23

**Table S3.** BECCS deployment levels in climate change mitigation scenarios from the AMPERE (available at: <https://tntcat.iiasa.ac.at/AMPEREDB/>)<sup>10,45</sup> and LIMITS (available at: <https://tntcat.iiasa.ac.at/LIMITSDB/>)<sup>36</sup> modelling comparison exercises. Only scenarios that apply the full unconstrained mitigation portfolio of the underlying models are shown (default technology assumptions). Policy scenarios include various short-term (i.e. including delayed action) and long-term climate targets as well as staged accession to an international climate agreement. The highest BECCS deployments are all produced by a single model (GCAM), which has the largest flexibility to compensate near term emissions by negative emissions in the second half of the century. BECCS deployment (1,4) is reported in negative values as carbon emissions are removed from the atmosphere. Gross emissions (2,6) could only be separated for carbon emissions from fossil fuel combustion and industry, but not for carbon emissions from land use which can include negative emissions from vegetation regrowth and afforestation. Therefore, net positive land use carbon emissions are included in gross emissions. All values converted from GtCO<sub>2</sub> to GtC and are rounded to two significant digits.

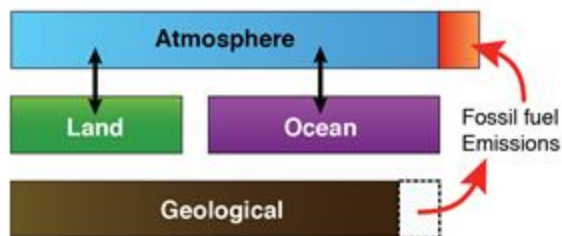
CO <sub>2</sub> eq concentration in 2100	<i>n</i>	Carbon emissions in 2100			Cumulative carbon emissions 2010-2100		
		BECCS deployment: amount of carbon removed (GtC/yr) (1)	Gross emissions (GtC/yr) (2)	Net total emissions (GtC/yr) (3)	BECCS deployment: amount of carbon removed (GtC) (4)	Gross emissions (GtC) (5)	Net total emissions (GtC) (6)
430-480	44	-3.3 (-5.9, -1.9)	1.8 (0.54, 2.3)	-2.6 (-5.9, -0.4)	-150 (-230, -100)	430 (390, 600)	280 (180, 320)
480-530	61	-4.1 (-15, -2.4)	2 (0.74, 3.3)	-2.7 (-16, 0.13)	-170 (-350, -87)	520 (480, 680)	320 (280, 460)
530-650	54	-3.4 (-15, 0)	3.3 (1.8, 4.6)	-1.1 (-16, 3.8)	-130 (-360, 0)	700 (620, 820)	560 (320, 690)

**Table S4.** Annual energy investments for BECCS technologies in 2030 and 2050 in a scenario consistent with staying below 2° C temperature rise. Data source: six global IAMs used in the LIMITS model inter-comparison (LIMITS-RefPol-450 scenario<sup>90</sup>). Average values across models are shown for each region, with full model ranges in parentheses. Investments into biofuels production with CCS and hydrogen production w/ CCS were not explicitly reported by modelling teams in the LIMITS exercise. Values for the former can be back-calculated, however, by multiplying total investments into biofuels production (both with and without CCS) and the share of total biofuels production that is equipped with CCS. While not exact, this estimation works quite well because the unit-level investment cost of a given biofuels production facility with CCS is only slightly greater than one without CCS. Totals may not add exactly due to rounding and averaging and because of a heterogeneous “REST\_WORLD” region that is not shown here. For regional definitions, see<sup>90</sup>.

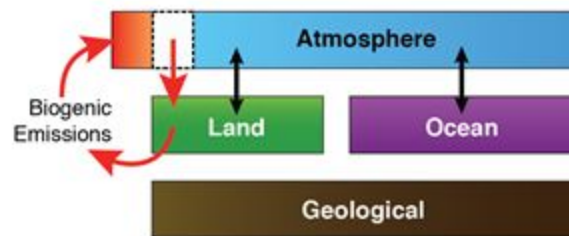
Investments into BECCS in a Scenario Consistent with 2° C				
	Biomass Electricity with CCS		Biofuels Production with CCS	
<i>units: billion US\$2005/yr</i>	2030	2050	2030	2050
<b>AFRICA</b>	1.2 (0 – 4.7)	17.2 (0 – 67.2)	0.5 (0 – 1.6)	8.1 (0 – 24.0)
<b>CHINA+</b>	2.9 (0 – 10.1)	30.5 (0 – 166.5)	6.4 (0 – 30.2)	16.4 (0 – 73.2)
<b>EUROPE</b>	3.8 (0 – 8.3)	12.4 (0 – 32.5)	3.6 (0 – 13.8)	9.4 (0 – 41.8)
<b>INDIA+</b>	4.5 (0 – 12.5)	10.4 (0 – 37.8)	4.6 (0 – 21.2)	6.9 (0 – 30.0)
<b>LATIN_AM</b>	1.7 (0 – 2.9)	15.8 (0 – 32.5)	2.4 (0 – 11.0)	17.5 (0 – 70.2)
<b>MIDDLE_EAST</b>	0.3 (0 – 1.1)	2.6 (0 – 12.1)	0.7 (0 – 1.9)	20.4 (0 – 100.4)
<b>NORTH_AM</b>	5.9 (0 – 21.6)	15.9 (0 – 39.9)	5.9 (0 – 28.8)	18.6 (0 – 74.9)
<b>PAC_OECD</b>	0.7 (0 – 2.2)	3.9 (0 – 10.2)	0.4 (0 – 1.8)	4.4 (0 – 11.4)
<b>REF_ECON</b>	2.8 (0 – 9.1)	10.9 (0 – 20.5)	2.1 (0 – 9.0)	3.2 (0 – 10.7)
<b>REST_ASIA</b>	5.6 (0 – 23.5)	11.9 (0 – 44.2)	2.1 (0 – 9.5)	10.4 (0 – 41.9)
<b>Developing</b>	16.2 (0 – 43.5)	88.3 (0 – 287.7)	16.6 (0 – 75.4)	79.7 (0 – 339.8)
<b>Industrialized</b>	20.0 (0 – 79.4)	50.0 (0 – 101.7)	12.7 (0 – 55.1)	43.0 (0 – 169.5)
<b>World</b>	36.2 (0 – 122.9)	138.3 (0 – 389.4)	29.4 (0 – 130.5)	122.6 (0 – 509.2)

**Figure S1.** Summary of drivers of and limits to the supply of NETs. Outward-pointing arrows represent activities that may increase the availability of NETs. Inward-pointing arrows represent key biophysical, economic, societal and climate-related limits to the global supply of NETs.

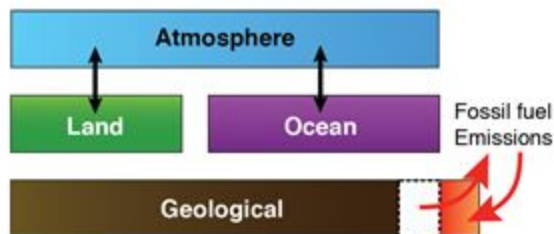
### A Fossil Fuel Energy



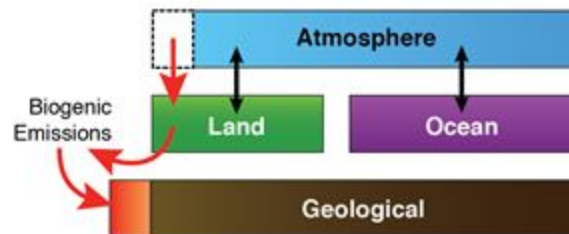
### B Bioenergy



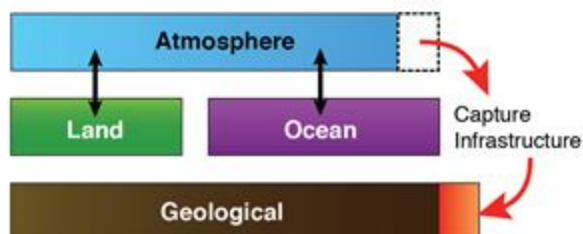
### C Carbon Capture & Storage (CCS)



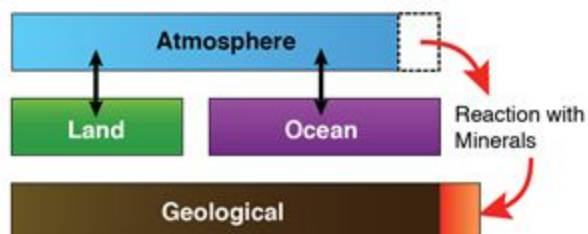
### D Bioenergy + CCS (BECCS)



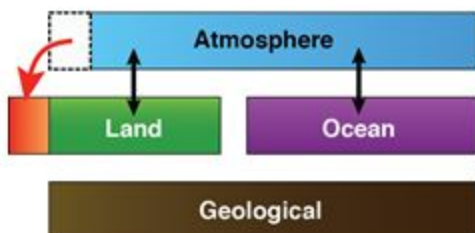
### E Direct Air Capture (DAC)



### F Enhanced Weathering



### G Afforestation/Changed Agricultural Practices



### H Ocean Fertilization/Alkalinization

